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THE EXTENDED RAYLEIGH THEORY OF THE OSCILLATION OF LIQUID DROPL--ETC(U)
MAY 80 C A MORRISON, R P LEAVITT, D E WORTMAN
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The Rayleigh theory of oscillation of liquid drops has been extended to include the case in which the drop is immersed in a uniform electric field. The resonant frequencies of the modes of the drop are shown to be shifted by the electric field. The magnitude and the direction of the shift are dependent on the dielectric constant of the drop. The condition for instability of drops in large electric fields is given and found to differ

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20. Abstract (Cont'd)

from that found by previous workers. This difference is attributed to the assumptions by previous workers that the drops, under the influence of an electric field, distorted into ellipsoids of revolution about the field direction. The dynamical equations are derived, and the solution for small oscillation is given.

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1. INTRODUCTION

The first work on the dynamical theory of the oscillation of liquid drops appears to have been done by Lord Rayleigh.[1] In his investigation, Rayleigh considered small displacements from equilibrium under the action of forces due to surface tension (capillary forces) alone. Later, Lamb[2] included damping of the small oscillation of drops by the inclusion of internal viscous forces and showed that the rate of damping was dependent on the size of the liquid drops, becoming extremely large for very small drops. The possibility of instability of the lower modes of oscillation was first investigated by Rayleigh,[3] who showed that the resonance frequency of the nth mode of a charged drop was

$$\omega_n^2 = \frac{n(n-1)}{\rho a^3}\left[\gamma(n+2) - \frac{Q^2}{4\pi a^3}\right] \quad ,$$

where ρ is the density of the liquid drop, a is the equilibrium radius, γ is the surface tension, and Q is the total charge on the drop. Instability occurs when $\omega_n^2 \leq 0$; or when the charge $|Q| > \left[4\pi a^3\gamma(n+2)\right]^{1/2}$, then the nth mode is unstable; the lower modes become unstable for the smaller amount of charge. The condition for instability is sometimes written in terms of the electric field at the surface of the drop, $E = Q/a^2$, so that the condition expressed above is frequently given as $|E| > \left[4\pi(n+2)\gamma/a\right]^{1/2}$ and has been used as a starting point by later investigators.

Considerably later than the above work, the investigation of the dynamics of small droplets was begun by Thacher[4] and O'Konski and Thacher.[5] In their investigations, they considered the distortion of droplets by an electric field, assuming that the distorted drop was an ellipsoid of revolution about the direction of the electric field. They also discussed the possibility of enhancing the droplet's distortion by an alternating electric field, but failed to observe that the frequency of the electric field should be half that required for resonance of the drop. O'Konski and Harris[6] extended this work to include in their analysis the effect of electrical conductivity of both the surrounding medium and the drop. No consideration was given to the possible effect of conductivity on the damping. In their analysis, they found the rather surprising result that, under certain appropriate choices of conductivities, the equilibrium shape of the droplet remained a sphere. As in their previous investigation, O'Konski and Harris assumed that the droplet under the action of an electric field becomes an ellipsoid of revolution about the field direction.

Later, Garton and Krasucki[7] investigated the stability of bubbles in a static electric field and questioned the correctness of previous theoretical work on electrostriction. They showed actual photographs of bubbles in various stages of disintegration that vividly displayed the physical nature of bubbles breaking.

In a series of two papers, Taylor[8] discussed the stability of conducting drops in an electric field. He showed that it is necessary to introduce motion of the fluid inside a drop to attain the spherical (undistorted) solution of O'Konski and Harris.[6] Some experimental work was reported in Taylor's papers that agreed quite well with the theory. Sozou[9] later extended Taylor's theory to include time dependent electric fields. He gave a number of results in the form of equations with a few cases of numerical results, but further work seems necessary for comparison with experiment and with previous results.

Rosenkilde[10] investigated the stability of droplets in an electric field using methods of tensor calculus and Chandrasekhar's[11] virial method. He showed that under appropriate conditions at least three different equilibrium configurations can exist. He predicted instability only if the dielectric constant, ε, of the drop is greater than 20.801.

[1] *Lord Rayleigh, Proc. Roy. Soc. (London), 29 (1879), 71; The Theory of Sound, II, MacMillan Co., London (1896), Ch. XX.*

[2] *H. Lamb, Hydrodynamics, Dover Publications, Inc., New York (1932).*

[3] *Lord Rayleigh, Phil. Mag., 14 (1882), 184.*

[4] *H. Thacher, Jr., J. Phys. Chem., 56 (1952), 795.*

[5] *C. T. O'Konski and H. Thacher, Jr., J. Phys. Chem., 57 (1953), 955.*

[6] *C. T. O'Konski and F. E. Harris, J. Phys. Chem., 61 (1957), 1172.*

[7] *G. G. Garton and Z. Krasucki, Proc. Roy. Soc. (London), A280 (1964), 211.*

[8] *Sir Geoffrey Taylor, Proc. Roy. Soc. (London), A280 (1964), 383; A291 (1966), 159.*

[9] *C. Sozou, Proc. Roy. Soc. (London), A331 (1972), 263.*

[10] *C. E. Rosenkilde, Proc. Roy. Soc. (London), A312 (1969), 473.*

[11] *S. Chandrasekhar, Hydrodynamics and Hydromagnetic Stability, Clarendon Press, Oxford (1961).*

Most of the above work was devoted to the development of equilibrium distortion of a droplet under the action of various forces. None of these workers attempted to develop their results using the original techniques introduced by Rayleigh.[1] In fact, the technique used by Rayleigh in his discussion of a charged drop[3] was somewhat different than in his original paper. A detailed derivation of Rayleigh's result for a charged drop was given by Hendricks and Schneider[12] and later by Schneider[13] along with many applications of the results. Dissipation by viscosity was not considered in either of these investigations.

In our consideration here, we follow the spirit of the original investigation by Rayleigh. In section 2.1, we find the energy due to the surface tension, the kinetic energy of the fluid in the drop, and the Rayleigh dissipation function. From these quantities, we derive the generalized equations of motion by using the Lagrangian, as given by Rayleigh, which includes the results of Lamb.

In section 2.2, we include the terms involving the energy due to the electric field in the Lagrangian. We discuss this aspect in considerable detail since we have not been able to find any reference to an extension of Rayleigh's result that includes an electric field.

The remainder of the report is devoted to a discussion of the result of section 2.2. The results are used in the investigation of instability under the action of an electrostatic field as well as the dynamical behavior of drops under the action of a time varying field.

2. THEORY

2.1 Derivation of Rayleigh's Result

In the dynamical theory of the oscillation of a liquid drop, we assume that the radius, r, from the center of mass to a point on the surface of the drop can be expanded in a Legendre series as

$$r(\theta,t) = a_0(t) + \sum_k{}' a_k(t)\, P_k(\cos\theta) \quad . \quad (1)$$

The method then of solving any particular problem is to express the Lagrangian for that problem in terms of the variables $a_n(t)$ and treat these $a_n(t)$ as generalized coordinates to obtain the equations of motion. Such an expansion as equation (1) is possible if we assume that the droplet is symmetric about the z-axis. At this point, the z-axis can be chosen in any direction; but later, when we include the electromagnetic energy, the electric field will be assumed to be along this axis. We assume this symmetry throughout the discussion. The prime on the sum in equation (1) indicates that the lowest value of k in the sum is one.

With the assumption of incompressibility, the volume of the drop acts as a constraint on the $a_k(t)$ in equation (1). The volume of the drop is given by

$$V = \int_0^{2\pi} d\phi \int_0^{\pi} \sin\theta\, d\theta \int_0^{r(\theta,t)} x^2\, dx \quad . \quad (2)$$

Since we are assuming that the drop is symmetric about the z-axis, letting $\mu = \cos\theta$, we have

$$V = \frac{2\pi}{3} \int_{-1}^{1} r^3(\mu)\, d\mu \quad . \quad (3)$$

Using equation (1) and the relation

$$\int_{-1}^{1} P_\ell(\mu) P_{\ell'}(\mu)\, d\mu = 2\delta_{\ell\ell'}/(2\ell + 1) \quad ,$$

we obtain

$$V \simeq \frac{2\pi}{3}\left(2a_0^3 + 3a_0 \sum_k{}' \frac{2a_k^2}{2k+1}\right) \quad . \quad (4)$$

If we assume that a is the radius of the equilibrium sphere ($V = 4\pi a^3/3$), then

$$a_0 \simeq a\left(1 - \frac{1}{a^2}\sum_k{}' \frac{a_k^2}{2k+1}\right) \quad , \quad (5)$$

[1]*Lord Rayleigh, Proc. Roy. Soc. (London), 29 (1879), 71; The Theory of Sound, II, MacMillan Co., London (1896), Ch. XX.*

[3]*Lord Rayleigh, Phil. Mag., 14 (1882), 184.*

[12]*C. D. Hendricks and J. M. Schneider, Am. J. Phys., 31 (1963), 450.*

[13]*John Matthew Schneider, The Stability of Electrified Liquid Surfaces, Charged Particle Research Laboratory, Univer ty of Illinois, Urbana, CPRL-2-64 (5 March 1964). (NTIS 604431).*

which holds for terms through order a_k^2 with $k \geq 1$. Since we are interested only in terms of second order in a_k, we treat equation (5) and corresponding subsequent results as equalities.

The potential energy, U_s, of the drop due to the surface tension, γ, is the area of the drop multiplied by γ or

$$U_s = \gamma \int_0^{2\pi} d\phi \int r \sin\theta \, ds \quad , \tag{6}$$

where ds is the arc length in the surface given by

$$ds^2 = dr^2 + r^2 \, d\theta^2$$

or

$$ds = d\theta \left[r^2 + \left(\frac{dr}{d\theta}\right)^2 \right]^{1/2} \quad . \tag{7}$$

Using this result in equation (6), we can write

$$U_s = 2\pi\gamma \int_{-1}^{1} r \left[r^2 + (1 - \mu^2)\left(\frac{dr}{d\mu}\right)^2 \right]^{1/2} d\mu \; . \tag{8}$$

The second term in the square root in equation (8) is smaller than r, and the square root can be expanded and integrated to give

$$U_s = 4\pi\gamma \left[a^2 + {\sum_k}' \frac{(k-1)(k+2)a_k^2}{2(2k+1)} \right] , \tag{9}$$

where the constraint given in equation (5) has been used to obtain this result. The result given in equation (9) holds up to fourth order in the a_k.

To calculate the kinetic energy, T, we need to evaluate the volume integral

$$T = \frac{1}{2} \int \rho v^2 \, d\tau \quad , \tag{10}$$

with ρ the density (constant) and v the velocity of the fluid within the drop. We assume that the fluid is incompressible and that there are no sources or sinks; then

$$\nabla \cdot \vec{v} = 0 \quad . \tag{11}$$

If further we assume that

$$\nabla \times \vec{v} = 0 \quad , \tag{12}$$

then $\vec{v}$ can be derived from a potential function, ϕ_1, such that

$$\vec{v} = -\nabla \phi_1 \quad , \tag{13}$$

and from equation (11) we obtain

$$\nabla^2 \phi_1 = 0 \quad . \tag{14}$$

Thus, the velocity potential satisfies Laplace's equation, and the solution appropriate for our problem can be written

$$\phi_1 = \sum_n \beta_n r^n P_n(\cos\theta) \quad . \tag{15}$$

(Here r represents a point on the interior of the drop and becomes $r(\theta,t)$ at the surface.) Rather than use equation (15) in equation (10) directly, we use equation (13) in equation (10) to convert the volume *integral into a surface integral as*

$$T = \frac{1}{2} \rho \int \phi_1 (\nabla \phi_1) \cdot d\vec{\sigma} \quad . \tag{16}$$

We assume that the area element $d\vec{\sigma}$ is approximately along $\hat{r}$. (Actually, $d\vec{\sigma} = (\hat{r}\, r\, d\theta - \hat{\theta}\, dr) \times r \sin\theta \, d\phi$, where $\hat{r}$ and $\hat{\theta}$ are unit vectors along r and θ, but the correction is of higher order than we are considering here.) Under that assumption, we have

$$T = 2\pi\rho a^2 {\sum_k}' \frac{k a^{2k-1}}{2k+1} \beta_k^2 \quad . \tag{17}$$

The β_k in equations (15) and (17) can be evaluated by equating the velocity at the surface $(-\partial\phi_1/\partial r)$ with $\dot{r}$ calculated from equation (1) so that

$$\dot{a}_k = -k a^{k-1} \beta_k \quad . \tag{18}$$

Then equation (17) becomes

$$T = 2\pi\rho a^3 \sum_k{}' \frac{\dot{a}_k^2}{k(2k+1)} \quad . \qquad (19)$$

This result, equation (19), is valid only through terms of order $\dot{a}_k^2$. If the result given in equation (18) is carefully inspected in the relation

$$v_r = -\sum k\beta_k r^{k-1} P_k(\cos\theta)$$

and the r^{k-1} is expanded by using equation (1), correction terms occur in β_k. These terms give corrections to β_k^2 in the form $\dot{a}_k a_n$, thereby coupling the equations of motion of a_k in a complicated manner, which we ignore in the present investigation. However, any serious investigation of dynamic instability should include these terms because they are of third order in the a_k and can be important.

To derive the effect of viscosity on the equations of motion, we use Rayleigh's dissipative function,[14] R, instead of the procedure used by Lamb.[2] As will become apparent in the development, this method is much more consistent with the treatment presented here. A convenient form of the dissipative function given by Landau and Lifshitz[15] is

$$R = \frac{1}{2}\eta \int (\nabla v^2) \cdot d\vec{\sigma} \quad , \qquad (20)$$

where η is the viscosity. As in the kinetic energy, it is sufficient to assume that $d\vec{\sigma}$ is along $\vec{r}$. All of the quantities necessary to calculate equation (20) are given in equations (15) and (18) so that

$$R = 4\pi\eta a \sum_k{}' \frac{(k-1)\dot{a}_k^2}{k} \quad . \qquad (21)$$

The equation of motion for $a_n(t)$ can now be readily found by forming the Lagrangian from the results given in equations (9) and (19) ($L_1 = T - U_s$) and the dissipative function equation (21) to give

$$\frac{4\pi\rho a^3}{n(2n+1)}\ddot{a}_n + 8\pi\eta \frac{a(n-1)}{n}\dot{a}_n + \frac{4\pi\gamma(n-1)(n+2)}{2n+1} a_n = 0 \quad . \qquad (22)$$

Thus, if the viscosity vanishes ($\eta = 0$), we have Rayleigh's result,[1]

$$\omega_n^2(\eta = 0) = \frac{\gamma}{\rho a^3} n(n-1)(n+2) \quad . \qquad (22a)$$

If the viscosity is small enough, we obtain Lamb's result[2] for the decay time of the nth mode $[a_n \sim a_n(0)\, e^{-t/\tau_n}]$,

$$\tau_n = \frac{\rho a^2}{\eta(n-1)(2n+1)} \quad . \qquad (22b)$$

The decay, when present, shifts the resonant frequency so that

$$\omega_n^2 = \omega_n^2(\eta = 0) - \left(\frac{1}{\tau_n}\right)^2 \qquad (22c)$$

for $\omega_n^2 \geq 0$; otherwise, the drop does not oscillate. For the mode $n = 2$, the frequency $\upsilon_2 = \omega_2/2\pi$, τ_2, and the product $\upsilon_2\tau_2$ were calculated by O'Konski and Thacher[5] for 1-, 10-, 100-, and 1000-μm water droplets, where they used $\gamma = 72.0$ dynes/cm and $\eta = 0.884 \times 10^{-2}$ poise.

All of the results obtained above were previously derived by various techniques, and we have shown that they can all be obtained by methods compatible with Rayleigh's original approach to the problem. Also, as we have shown, corrections to Rayleigh's results are more transparent by this approach.

2.2 Electric Field

The inclusion of an electric field into the analysis presents a problem of considerable com-

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[2] *H. Lamb, Hydrodynamics, Dover Publications, Inc., New York (1932).*

[14] *H. Goldstein, Classical Mechanics, Addison-Wesley Publishing Co., Reading, MA (1950), Ch. 1.*

[15] *L. D. Landau and E. M. Lifshitz, Fluid Mechanics, Theoretical Physics, 6, Addison-Wesley Publishing Co., Reading, MA (1959), 54.*

[1] *Lord Rayleigh, Proc. Roy. Soc. (London), 29 (1879), 71; The Theory of Sound, II, MacMillan Co., London (1896), Ch. XX.*

[2] *H. Lamb, Hydrodynamics, Dover Publications, Inc., New York (1932).*

[5] *C. T. O'Konski and H. Thacher, Jr., J. Phys. Chem., 57 (1953), 955.*

plexity. Therefore, we present the approach and the solution to the problem in more detail than in our previous discussion. To be consistent with our preceding analysis, we need to obtain an expression for the electromagnetic energy of the drop expressed in terms of the $a_n(t)$ in equation (1). This energy can be added to the existing Lagrangian, which can then be used to obtain the equations of motion.

A convenient form for the electromagnetic energy stored in the drop is given by Jackson:[16]

$$U_E = -\frac{\varepsilon - 1}{8\pi} \int \vec{E}^{\,i} \cdot \vec{E}\, d\tau \quad , \qquad (23)$$

where $\vec{E}^{\,i}$ is the electric field inside the drop, $\vec{E}$ is the field in the absence of the drop, ε is the dielectric constant of the drop, and the integral covers the volume of the drop. Thus, our problem is to find a solution for $\vec{E}^{\,i}$ before proceeding.

For a dielectric body, we have from Maxwell's equations

$$\begin{aligned} \nabla \times \vec{E} &= 0 \quad , \\ \nabla \cdot \vec{D} = \varepsilon\, \nabla \cdot \vec{E} &= 0 \quad . \end{aligned} \qquad (24)$$

The first of equation (24) implies that the electric field can be derived from a scalar potential, ψ, or

$$\vec{E} = -\nabla\psi \quad , \qquad (25)$$

and the second of equation (24) implies that

$$\nabla^2\psi = 0 \quad . \qquad (26)$$

The appropriate solutions to equation (26) for our problem can be written

$$\psi^o = \sum_{n=0}^{\infty} \frac{A_n}{r^{n+1}} P_n(\cos\theta) - Er\cos\theta \qquad (27)$$

[16] J. D. Jackson, *Classical Electrodynamics*, John Wiley and Sons, New York (1975), 160. The derivation of equation (23) is not trivial, and Jackson gives considerable attention to the derivation.

for the exterior region and

$$\psi^i = \sum_n r^n B_n P_n(\cos\theta) \qquad (28)$$

for the interior region of the drop. The last term in equation (27) represents the potential of the external electric field, E, parallel to the z-axis.

The applied electric field, $\vec{E}$, can be written

$$\vec{E} = E(\hat{r}\cos\theta + \hat{\theta}\sin\theta) \quad , \qquad (29)$$

$\hat{r}$ and $\hat{\theta}$ are unit vectors in their respective direction, and from equation (25)

$$\vec{E}^{\,i} = -\nabla\psi^i \quad .$$

Then using equation (27), we have

$$\vec{E} \cdot \vec{E}^{\,i} = -E \sum_n B_n r^{n-1} \Big[n\cos\theta\, P_n(\cos\theta) - \sin\theta \frac{\partial}{\partial\theta} P_n(\cos\theta) \Big] \quad . \qquad (30)$$

From the recursion relation for Legendre polynomials, with $\mu = \cos\theta$, we have

$$(1 - \mu^2)\frac{dP_n}{d\mu} = nP_{n-1} - n\mu P_n \quad ,$$

where we have discontinued writing the argument in the Legendre polynomials. Using this result in equation (30), we have

$$\vec{E} \cdot \vec{E}^{\,i} = -E \sum_n nB_n r^{n-1} P_{n-1} \qquad (31)$$

and

$$\int_0^{2\pi} d\phi \int_0^{\pi} \sin\theta\, d\theta \int_0^{r(\theta,t)} r^2\, \vec{E} \cdot \vec{E}^{\,i}\, dr$$

$$= -2\pi E \sum_n \frac{n}{n+2} B_n \int_{-1}^{1} r^{n+2} P_{n-1}\, d\mu \quad .$$

Substituting this result into equation (23) we have

$$U_E = \frac{\varepsilon - 1}{4} E \Bigg[\frac{2B_1}{3} a^3 + \sum_{n>1} \frac{nB_n}{n+2}$$

$$\times \int_{-1}^{1} r^{n+2} P_{n-1}\, d\mu \Big] \quad , \qquad (32)$$

where we have separated the term $n = 1$ from the remainder since this term is the integral over μ of r^3, which is proportional to the volume and is the constraining equation (3).

To evaluate the terms in the sum in equation (32), we use equation (1) and expand the powers of a_0 through linear terms in a_k or

$$\int_{-1}^{1} r^{n+2} P_{n-1}\, d\mu \simeq \int_{-1}^{1} \Big[a_0^{n+2} + (n+2) a_0^{n+1} \sum_k{}' a_k P_k \Big] P_{n-1}\, d\mu$$

$$= \frac{2(n+2)}{(2n-1)} a_0^{n+1} a_{n-1} \qquad (33)$$

for $n > 1$. The resulting equation (33) can be used in equation (32) to give

$$U_E = \frac{\varepsilon - 1}{2} E \left[\frac{B_1}{3} a^3 + \sum_{n=1}^{\infty} \frac{n+1}{2n+1} B_{n+1} a^{n+2} a_n \right] \quad , \quad (34)$$

and we have shifted the summing index so that the lower limit on n is 1. We have also replaced the product $a_n a_0^{n+2}$ by $a_n a^{n+2}$ since the corrections to a_0 are of order a_k^2 so that corrections to this quantity would be of order a_k^3, which are neglected. Also, in equation (33), we have expanded r only through terms linear in a_k because B_n in equation (34) for $n > 1$ have to depend on a_k since they vanish for undistorted spherical drops. (That they vanish appears in the subsequent analysis.) Therefore, to obtain an expression valid through terms of order a_k^2, it is necessary to obtain B_1 through terms of order a_k^2 for contributions from the first term in equation (34), while we need only first order terms for B_n with $n > 1$. To proceed, we have to return to the problem of evaluating the B_n and consequently the A_n of equations (27) and (28).

To evaluate B_n and A_n in equations (27) and (28), we need to apply the boundary conditions on the fields at the surface of the drop. These are that the tangential components of the electric field are continuous, $E_t^i = E_t^o$, and the normal components of the electric displacement are continuous, $D_n^i = D_n^o$, where the subscripts t and n on the vector components denote tangential and normal, respectively. (The use of n here and in the summing index need cause no confusion since no sums are performed over the field components.) The continuity of the tangential electric field can be easily shown to be equivalent to the continuity of the potential so that $\psi^i = \psi^o$ at the surface. The boundary condition on the normal component of $\vec{D}$, however, requires some consideration. Since by equation (25) $\vec{E} = -\nabla \psi$ and since we are assuming that $\vec{D}^i = \varepsilon \vec{E}^i$ and $\vec{D}^o = \vec{E}^o$ with $\hat{n}$ a unit vector normal to the surface, we have

$$\varepsilon\, \hat{n} \cdot \nabla \psi^i = \hat{n} \cdot \nabla \psi^o \qquad (35)$$

at the boundary of the drop. To construct $\hat{n}$, we note that the vector $\vec{ds} = \hat{r}\, dr + \hat{\theta} r\, d\theta$ lies in the surface if r is given by equation (1) and the unit vector $\hat{\phi}$ in the ϕ direction also lies in the surface since we are assuming that the drop is symmetrical about the z-axis. A vector normal to the surface can be formed by the cross product of these two vectors or

$$\hat{n} = \frac{\vec{ds} \times \hat{\phi}}{|\vec{ds} \times \hat{\phi}|}$$

$$= \frac{\hat{r}\, r\, d\theta - \hat{\theta}\, dr}{(r^2\, d\theta^2 + dr^2)^{1/2}} \quad , \qquad (36)$$

which can be used in equation (35) to obtain the normal derivative. Thus, to reiterate, the boundary conditions on equations (27) and (28) are

$$\psi^o = \psi^i \quad ,$$

$$\varepsilon \left[\frac{\partial \psi^i}{\partial r} - \left(\frac{dr}{r\, d\theta} \right) \frac{\partial \psi^i}{r\, \partial \theta} \right] = \frac{\partial \psi^o}{\partial r} - \left(\frac{dr}{r\, d\theta} \right) \frac{\partial \psi^o}{r\, \partial \theta} \qquad (37)$$

evaluated at the boundary with r given by equation (1).

To keep track of the order of the corrections, it is convenient to rewrite equation (1) as

$$r = a_0 + \delta \sum{}' a_k P_k$$

and expand the various powers of r in powers of δ; also, we assume that

$$A_n = A_n^{(0)} + \delta A_n^{(1)} + \delta^2 A_n^{(2)} \ldots \quad ,$$

$$B_n = B_n^{(0)} + \delta B_n^{(1)} + \delta^2 B_n^{(2)} \ldots \quad . \qquad (38)$$

Then in the final result, we let $\delta = 1$ since it serves only as an artifice to keep the terms in order. When this is done in equation (38), we obtain

$$A_n^{(0)} = B_n^{(0)} = 0 \ , \quad n \neq 1 \quad ,$$

$$A_1^{(0)} = \frac{(\varepsilon - 1)Ea_0^3}{\varepsilon + 2} \quad ,$$

$$B_1^{(0)} = -\frac{3E}{\varepsilon + 2} \quad ,$$

$$A_n^{(1)} = \frac{3(\varepsilon - 1)E}{\varepsilon + 2}\left\{\frac{n(n+1)(\varepsilon - 1)a_{n+1}}{(2n+3)[(\varepsilon+1)n+1]} + \frac{na_{n-1}}{2n-1}\right\}a_0^{n+1} \quad ,$$

$$B_n^{(1)} = -\frac{3(\varepsilon - 1)E}{\varepsilon + 2}\left\{\frac{(n+1)(2n+1)a_{n+1}}{(2n+3)[(\varepsilon+1)n+1]a_0^n}\right\} ,$$

and

$$B_1^{(2)} = -\frac{9(\varepsilon - 1)E}{(\varepsilon + 2)^2 a_0^2} \times \sum_m \left(G_m a_m a_{m-2} + H_m a_m^2\right) \quad , \qquad (39)$$

where

$$G_m = \frac{m(m-1)(2m-1)(\varepsilon+2)}{(2m-3)(2m+1)[\varepsilon(m-1)+m]} \quad ,$$

$$H_m = \frac{m[\varepsilon(m-1)(12m^3 + 10m^2 - 12m - 1)]}{(2m+1)^2(2m-1)(2m+3)[\varepsilon(m-1)+m]} - \frac{m^2(12m^3 + 2m^2 - 18m + 10)}{(2m+1)^2(2m-1)(2m+3)[\varepsilon(m-1)+m]} \ .$$

When the results given in equation (39) are substituted into equation (34), we have

$$U_E = -\frac{(\varepsilon - 1)E^2 a}{2(\varepsilon + 2)}\left[a^2 + \frac{6(\varepsilon - 1)a}{5(\varepsilon + 2)}a_2 + \frac{6(\varepsilon - 1)}{\varepsilon + 2}\sum_{m=1}^{\infty}\left(G_{m+2}a_m a_{m+2} + H_m a_m^2\right)\right] \ . \qquad (40)$$

To form the full Lagrangian for the problem, we need only add U_E to U_s so that $L = T - U_s - U_E$, and the equation of motion for a_n is given by the usual procedure,

$$\frac{d}{dt}\frac{\partial L}{\partial \dot{a}_n} - \frac{\partial L}{\partial a_n} = \frac{\partial R}{\partial a_n} \ . \qquad (41)$$

The result of doing this is to add to the right side of equation (22) the term $-\partial U_E/\partial a_n$ so that

$$\frac{4\pi\rho a^3}{n(2n+1)}\ddot{a}_n + 8\pi\eta\frac{a(n-1)}{n}\dot{a}_n + \frac{4\pi\gamma(n-1)(n+2)}{2n+1}a_n = \frac{3(\varepsilon - 1)^2E^2a}{5(\varepsilon+2)^2}\delta_{n,2} + \frac{3(\varepsilon-1)^2E^2a}{(\varepsilon+2)^2} \times \left(G_{n+2}a_{n+2} + G_n a_{n-2} + 2H_n a_n\right), \quad (42)$$

which are the equations of motion for a drop in an electric field. The right hand side of equation (42) appears to be new.

Several aspects of equation (42) are as should be expected from the general symmetry of the problem. That is, the right hand side is dependent on the square of the electric field so that a reversal of the field does not alter the results. Further, the $n = 2$ mode is the only mode driven directly by the electric field, and this couples to the $n = 4$ mode and consequently to higher modes with n even. Consequently if no further perturbation couples the odd and even modes, only

those modes for n even need to be considered. Some of these results would have been immediately obvious if we had used Maxwell's stress tensor to evaluate the forces on the drop, but then the normal mode result of equation (42) would have been lost. If we ignore the coupling to the higher modes in equation (42), the resonant frequency of the nth mode is given by

$$\omega_n^2 = \frac{n(n-1)}{\rho a^3} \times \left[\gamma(n+2) - \frac{6(\varepsilon - 1)^2 E^2 a(2n+1)H_n}{4\pi(n-1)(\varepsilon+2)^2} \right] . \tag{43}$$

This result is similar in appearance to the result derived by Rayleigh for a charged drop,

$$\omega_n^2 = \frac{n(n-1)}{\rho a^3}\left[\gamma(n+2) - \frac{E^2 a}{4\pi}\right] ,$$

with the charge on the drop, $Q = Ea^2$. In Rayleigh's result, the frequency decreases as the *field (charge) increases, but in equation (43) this* condition occurs only if $H_n > 0$. The condition for no shift in the frequency in equation (43) is $H_n = 0$, which for the first few modes are $n = 2$, $\varepsilon = 2.846$; $n = 4$, $\varepsilon = 1.588$; $n = 6$, $\varepsilon = 1.346$; and $n = 8$, $\varepsilon = 1$. Perhaps a fair comparison with Rayleigh's result is for a conducting drop, which can be obtained from equation (43) by letting the dielectric constant be infinite or

$$\omega_n^2(\varepsilon = \infty) = \frac{n(n-1)}{\rho a^3} \times \left[\gamma(n+2) - \frac{6E^2 a}{4\pi} \times \frac{n\left(12n^3 + 10n^2 - 12n - 1\right)}{(n-1)(2n+1)(2n-1)(2n+3)}\right] . \tag{44}$$

This resulting equation (44) shows the same behavior as the result of Rayleigh, but with a more complicated dependence on mode number, n.

3. DISCUSSION

The result given in equation (42) is to this point general in that no assumptions have been made concerning the nature of the electric field other than those implied in equations (24) and (25). In this section, we discuss results from equation (42) for three different types of fields: a static electric field, an alternating electric field, and an amplitude modulated high frequency field.

3.1 Static Electric Field

In the case of a charged drop under the influence of its self-electric field, the modes of oscillation are uncoupled so that a simple criterion, $\omega_n = 0$, is usually taken as the onset of instability. In the present case given in equation (42), we see that the modes are coupled so that the condition for instability created by a large electric field becomes complicated. If we assume that $\ddot{a}_n$ and $\dot{a}_n$ in equation (42) vanish, then we can write

$$D_n x_n = \frac{3}{5}\lambda y \delta_{n,2} + 3\lambda y\left(G_{n+2}x_{n+2} + G_n x_{n-2}\right) , \tag{45}$$

where

$$D_n = A_n - 6\lambda y H_n ,$$

$$A_n = \frac{4\pi(n-1)(n+2)}{2n+1} ,$$

$$x_n = \frac{a_n}{a} ,$$

$$\lambda = \left(\frac{\varepsilon - 1}{\varepsilon + 2}\right)^2 ,$$

$$y = \frac{E^2 a}{\gamma} ,$$

with G_n and H_n given in equation (39). For low fields ($y \sim 0$), the coupling between modes can

generally be ignored; but for larger fields, which would be required for the drop to become unstable, the coupling cannot be ignored. Thus to investigate the instability in the system of equations in equation (45), it is necessary to consider the coupled equations in detail.

If we write equation (45) in matrix notation, then

$$T\vec{x} = \vec{F} \quad , \tag{46}$$

where

$$T_{n'n} = D_n\delta_{n'n} - 3\lambda G_{n'}\delta_{n',n+2} - 3\lambda G_n\delta_{n',n-2}$$

and $\vec{x}$ and $\vec{F}$ are column matrices (vectors). The matrix $\vec{F}$ consists of the single element, $(3/5)\lambda y$. The formal solution to equation (46) is given by

$$\vec{x} = T^{-1}\vec{F} \quad , \tag{47}$$

where T^{-1} is the inverse of the matrix T. The inverse of the matrix T contains the determinant of T in the denominator, and when the determinant vanishes, the solution equation (47) becomes unstable. Thus the lowest value of electric field (smallest positive y) for which the determinant vanishes gives the onset of instability of the drop. Since the matrix T has single off-diagonal elements, it is a simple matter to obtain a recursion relation for the determinant, Δ. If the matrix is truncated to contain N terms ($T_{nn'} = 0$, n or $n' > 2N$), then

$$\Delta_{2N} = D_{2N}\Delta_{2N-2} - (3\lambda y G_{2N})^2\Delta_{2N-4} \quad , \tag{48}$$

with $\Delta_0 = 1$ and $\Delta_{2N-2} = 0$ for N negative. The result given in equation (48) can be used to obtain the smallest positive value of y such that $\Delta_{2N}(y) = 0$. The first two of these are from equation (48),

$$\Delta_2(y) = A_2 - 6\lambda y H_2 \quad , \quad \Delta_2(y) = 0 \text{ gives}$$

$$y_2 = \frac{A_2}{6\lambda H_2} \tag{49}$$

and

$$\Delta_4(y) = (A_4 - 6\lambda y H_4) \times (A_2 - 6\lambda y H_2) - (3\lambda y G_4)^2 \quad , \tag{50}$$

$$y_4 = \frac{H_4A_2 + H_2A_4}{3\lambda(4H_4H_2 - G_4^2)} - \frac{[(H_4A_2 - H_2A_4)^2 + A_2A_4G_4^2]^{1/2}}{3\lambda(4H_4H_2 - G_4^2)} \quad ,$$

which are the simplest roots to obtain algebraic expressions for. The ambiguity of sign in equation (50) was determined so that, y_4 given, there was the smallest positive root in the range of dielectric constants, $1 \le \varepsilon \le \infty$. The result for y_2 in equation (49) is not physical ($y_2 < 0$) for $\varepsilon \le 52/37$ and becomes infinite at $\varepsilon = 52/37$ (H_2 vanishes) and is, in general, not even a good first estimate. This result is not surprising since y_2 totally ignores the coupling between modes. The result for y_4 given in equation (50) is, on the other hand, a good estimate even for small values of ε. The lowest values of y for which $\Delta_{2N}(y) = 0$ have been calculated for a few values of N and an extended range of ε and are given in table 1. The value of y_4 differs from the value of y_{10} only by 16 percent for $\varepsilon = 1.1$; for water ($\varepsilon = 78.2$), y_4 differs from y_{10} by 1.3 percent, and for larger ε the difference is approximately 1 percent.

TABLE 1. CRITICAL VALUES OF y (E^2a/γ) ARE LOWEST VALUES OF y FOR WHICH $\Delta_k(y_k) = 0$ FOR k = 4, 6, 8, AND 10

ε	y_4	y_6	y_8	y_{10}
1.1	7009	6109	6054	6049
1.3	817.6	728.2	720.5	720.0
1.5	310.9	280.4	278.2	278.1
2.0	90.16	83.71	82.80	82.78
5.0	12.87	12.42	12.40	12.39
10.0	6.511	6.367	6.362	6.362
50.0	3.603	3.556	3.555	3.555
78.2	3.406	3.364	3.363	3.363
100	3.332	3.302	3.291	3.291
∞	3.078	3.044	3.044	3.044

Upon comparing the results in table 1 with the results obtained by Brazier-Smith et al,[17] we find a discrepancy. The value of $E(a/\gamma)^{1/2} = 1.625$ ($y = E^2a/\gamma$) given by them (along with references to previous work) corresponds to our value of 1.745 for $\varepsilon = \infty$. For water, which is used in their experiment, we obtain 1.834. This discrepancy is caused possibly by the assumption that the drop is constrained to an ellipsoid of revolution in their treatment of the problem.

The amplitude of the x_2 mode is easily obtained by using equation (45) along with the determinant in equation (48) to give for the 2Nth approximate solution

$$x_2(2N) = \frac{P_{2N}}{\Delta_{2N}} \quad , \qquad (51)$$

where the P_{2N} obey the same recursion relation as Δ_{2N} given in equation (48). The initial values of P_{2N} are

$$P_2 = 3\lambda y/5 \qquad (52)$$

and

$$P_4 = 3\lambda y D_4/5 \quad , \qquad (53)$$

which are sufficient to generate all P_{2N} from equation (48). Having determined the value of $x_2(2N)$, we can generate *all* the $x_n(2N)$ for $n \leq 2N$ by using equation (45) and the condition $x_0 = 0$. The variation of $x_2(10)$ as a function of y is shown in figures 1 and 2 for two values of dielectric constant, $\varepsilon = 78.2$ and $\varepsilon = \infty$. The amplitudes of the x_n mode are approximately an order of magnitude larger than the x_{n+2} mode in the moderate y region. All the modes, x_n, diverge at the smallest value of y, satisfying the equation $\Delta_{10}(y) = 0$. When the x_n become large, the results are questionable, and a possible measure of the range of x_n that are reasonable is from equation (5) (with $x_n = a_n/a$),

$$\frac{a_0}{a} = 1 - \sum_{n=1}^{\infty} \frac{x_n^2}{2n+1} \quad . \qquad (54)$$

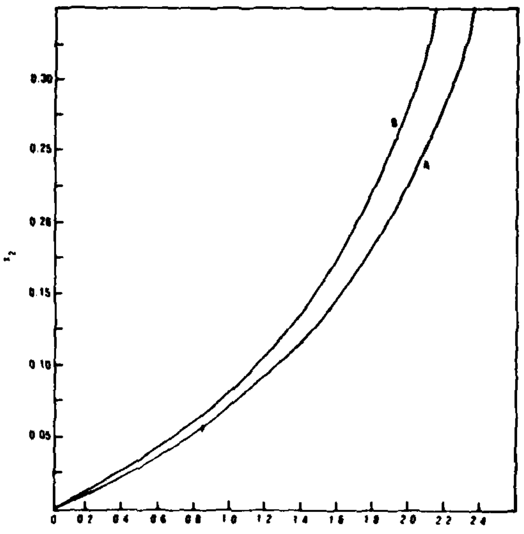

Figure 1. Amplitude of second mode, x_2, as function of y (E^2a/γ) for dielectric constant $\varepsilon = 78.2$ (A) and $\varepsilon = \infty$ (B).

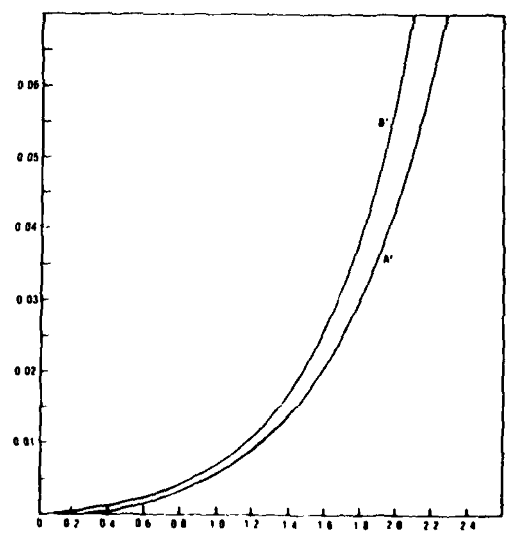

Figure 2. Amplitude of fourth mode, x_4, as function of y (E^2a/γ) for dielectric constant $\varepsilon = 78.2$ (A') and $\varepsilon = \infty$ (B').

[17] *P. R. Brazier-Smith, M. Brook, J. Latham, C.P.R. Saunders, and M. H. Smith, Proc. Roy. Soc. (London), A322 (1971), 523.*

When a_0/a becomes small, the entire results become questionable because all the expansions used in the derivation equation (45) assume $a_0 > |a_k|$ implicitly.

If the drop were distorted in the shape of an ellipsoid of revolution along the direction of the field, we could write

$$\frac{r^2 \sin^2 \theta}{A_1^2} + \frac{r^2 \cos^2 \theta}{B_1^2} = 1 \quad . (55)$$

If we let $\theta = \pi/2$, we have from equation (1)

$$A_1 = r\left(\frac{\pi}{2}\right) = \sum a_n P_n(0) \quad (56)$$

or

$$A = \frac{a_0}{a} + \sum_{n=1}^{\infty} x_{2n}(-1)^n \binom{2n}{n} \frac{1}{2^{2n}} \quad , \quad (57)$$

where $A = A_1/a$

and $\binom{2n}{n}$ is a binomial coefficient.

If we let $\theta = 0$ or π in equation (55), then from equation (1)

$$B_1 = \begin{cases} r(\theta = 0) = a_0 + \sum' a_n \quad , \\ r(\theta = \pi) = a_0 + \sum' (-1)^n a_n \quad . \end{cases}$$

Then

$$B = \frac{a_0}{a} + \sum' x_{2n} \quad . \quad (58)$$

where $B = B_1/a$. If the distortion were an ellipsoid of revolution as given in equation (55), the volume of the drop would be given by $A^2B = 1$. For $\varepsilon = \infty$, a_0/a was computed from equation (54), A by equation (57), and B by equation (58) as a function of y, and the results are shown in figure 3. A measure of how closely the drop approximates an ellipsoid of revolution is $1 - A^2B$; for small values of y, this quantity is quite small. However, for $y = 2.8$, $1 - A^2B \sim 0.28$, and a_0/a is still large (~ 0.9). The ellipsoidal approximation, when viewed in this light, would not seem very good and perhaps would indicate the source of the difference in the critical field values obtained here and elsewhere by earlier workers. (See fig. 3 on p. 16.)

3.2 Dynamical Equation for Varying Electric Fields

The dynamical equation (42) is, to our knowledge, a new result in the sense that the losses due to viscosity and a finite dielectric constant have been included in the analysis. Further, the entire equation of motion was derived in a consistent manner by using Rayleigh's original method. For convenience, we can write equation (42) in the form

$$M_n\ddot{x}_n + M_n\Gamma_n\dot{x}_n + M_n\omega_n^2 x_n = \frac{3}{5}\lambda E^2 a\delta_{n,2} + 3\lambda E^2 a\left(G_{n+2}x_{n+2} + G_n x_{n-2}\right) \quad , \quad (59)$$

where

$$M_n = \frac{4\pi\rho a^3}{n(2n+1)}$$

(M_1 is the total mass of the drop),

$$\Gamma_n = \frac{2\eta a(n-1)(2n+1)}{\rho a^3} \quad ,$$

$$\omega_n^2 = \omega_n^2(0) - \frac{2\lambda E^2 a}{M_1} n(2n+1)H_n \quad ,$$

where $\omega_n^2(0)$ is the frequency in the absence of an electric field given in equation (22a) and H_n is given in equation (39). The quantity λ is defined in equation (45). If the electric field is assumed in the form

$$E = E_0 \cos \omega t \quad , \quad (60)$$

then

$$E^2 = \frac{E_0^2}{2} + \frac{E_0^2}{2} \cos 2\omega t \quad . \quad (61)$$

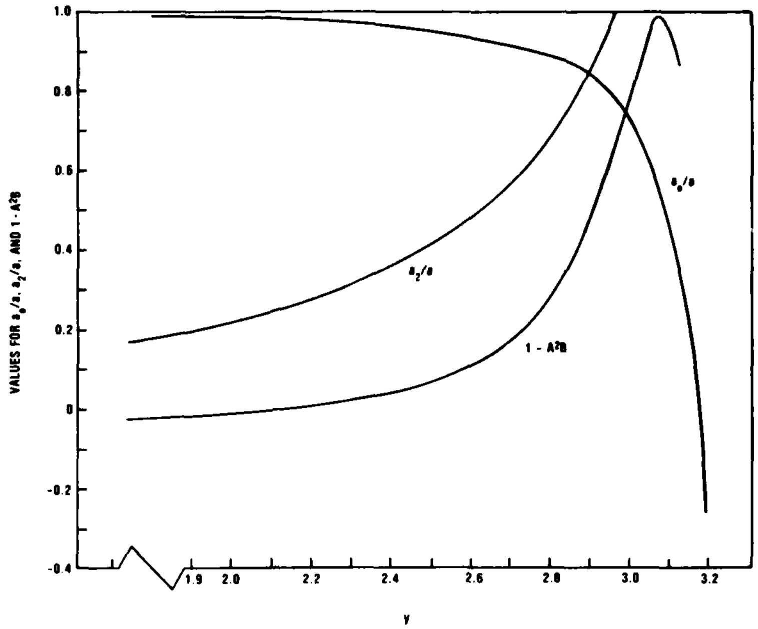

Figure 3. Quantities a_0/a, a_2/a, and $1 - A^2B$ obtained from normal mode analysis versus field strength y.

Thus, the driving force in equation (59) for the $n = 2$ mode varies at twice the frequency of the electric field, and resonance for small amplitude of oscillation should occur near $2\omega = \omega_n$.

If we ignore the coupling to higher modes ($x_0 = 0$) in equation (59) and replace E^2 by $E_0^2/2$ in ω_n^2, we have

$$\ddot{x}_2 + \Gamma_2 \dot{x}_2 + \omega_2^2 x_2$$

$$= \frac{3\lambda E_0^2 a}{10M_2} + \frac{3\lambda E_0^2 a}{10M_2} \cos 2\omega t \quad , (62)$$

where we have used the results of equation (61). The result given in equation (62) is a standard linear equation, and the solution can be written $x_2(t) = x_2^0 + \xi_2(t)$ with

$$x_2^0 = \frac{3\lambda E_0^2 a}{10M_2\omega_2^2}$$

and

$$\xi_2(t) = \frac{3\lambda E_0^2 a}{10M_2R_2} \cos (2\omega t - \theta_2) \quad , \quad (63)$$

where

$$R_2 = \left[\left(\omega_2^2 - 4\omega^2\right)^2 + 4\omega^2\Gamma_2^2\right]^{1/2}$$

and

$$\tan \theta_2 = \frac{\omega\Gamma_2}{\omega_2^2 - 4\omega^2} \quad .$$

Also, in the derivation of equation (63), we have ignored terms of higher power than E_0^2 in the electric field.

Thus the results given in equation (63) show that in small electric fields the drop, in the fundamental mode, is similar to an ordinary reso-

nant circuit with damping; however, the driving field on the drop has a frequency double that of the applied field. The higher modes are driven by the electric field indirectly by coupling through the lower modes as shown in equation (59), and approximate solutions can be obtained by a perturbation theory solution of the equations of motion. If the coupling is ignored, the amplitude of vibration of the nth mode can be written

$$x_n(t) = a_n^{(0)} e^{-\Gamma_n/2} \cos\left(\Omega_n t + \alpha_n\right) \qquad (64)$$

for

$$\Gamma_n^2/4 < \omega_n^2$$

and

$$\Omega_n = \left(\omega_n^2 - \frac{\Gamma_n^2}{4}\right)^{1/2} ,$$

with ω_n and Γ_n given by equation (59). The result in equation (64) shows that the nth mode decays more rapidly than the fundamental ($n = 2$); that is, since

$$\Gamma_n = \frac{(n-1)(2n+1)}{5} \Gamma_2 \quad ,$$

the $n = 4$ mode decays approximately six times faster than the fundamental. The shift in frequency due to viscosity and electric field can be written

$$\Delta\omega_n = \omega_n(0) - \Omega_n \quad . \qquad (65)$$

Assuming the electric field and the viscosity small, this can be written

$$\frac{\Delta\omega_n}{\omega_n(0)} = \left[\frac{\Gamma_n^2}{8\omega_n^2(0)} + \frac{\lambda E^2 a}{M_1\omega_n^2(0)} n(2n+1)H_n\right] . \qquad (66)$$

In general, the shift in frequency given in equation (66) is positive. However, H_n given in equation (39) can be positive or negative depending on E so that in certain cases the resonant frequency could be unshifted if the viscosity were small enough. Care must be exercised in extending equation (66) to higher electric fields because the coupling to higher modes given in equation (59) can have more important consequences. Also, for higher electric field strengths, the term in ω_n^2 in equation (59) involving the electric field has a time dependent part that could have some interesting consequences in large electric fields.

The dynamical equation (62) or the more general equation (59) can be used in more complicated situations where the electric field is of an impulse nature such as that caused by the presence of charged drops. A number of sources of electrical and mechanical disturbances have been considered by Brook and Lantham in their study of modulation of radar echo from rainstorms.[18]

A second and perhaps a more interesting case of varying fields is the electric field as an amplitude modulated high frequency wave. The carrier may be radar or an infrared laser, and the amplitude modulation can be chosen near a resonance of the drop. Such a field can be represented by

$$E = E_0(1 + m_0 \cos \omega t) \cos \omega_0 t \quad , \qquad (67)$$

where $\omega_0 >> \omega$, and particular space dependence has been ignored because we assume that the drop radius is small compared with the wavelength, λ_0, of the carrier ($\lambda_0 = 2\pi c/\omega_0$). We need the square of the field, which is given by

$$E^2 = \frac{E_0^2}{2}\left(1 + \frac{m_0^2}{2} + 2m_0 \cos \omega t + \frac{m_0^2}{2} \cos 2\omega t\right) . \qquad (68)$$

If we ignore all the high frequency terms varying at frequencies near $2\omega_0$ and if the depth of modulation m_0 is not large, the terms involving m_0^2 can

[18] M. Brook and D. J. Lanthan, J. Geophys. Res., 73 (1968), 7137.

be ignored. Substituting equation (68) into equation (59), we obtain

$$\ddot{x}_n + \Gamma_n\dot{x}_n + \omega_n^2 x_n = \frac{3\lambda E_0^2 a}{10}$$

$$\times (1 + 2m_0 \cos \omega t)\delta_{n2} + \frac{3\lambda E_0^2 a}{2}$$

$$\times (1 + 2m_0 \cos \omega t)(G_{n+2}x_{n+2} + G_n x_{n-2}) \ , \quad (69)$$

where as in equation (62) we assume that the term E^2 in ω_n^2 is replaced by $E_0^2/2$. Also implicit in the equation is the assumption that the dielectric constant, ε, wherever it appears, is to be evaluated at the carrier frequency, ω_0. Only in cases of extreme dispersion is this invalid, and such cases have to be treated quite differently. The solutions to equation (69) are the same as in equation (62) for $n = 2$, except that the frequency here is at half the value used in equation (62).

The time varying displacement of the drops could be studied[19] by amplitude modulating a high powered laser and observing the modulation of the reflected signal by a second low power laser. Some of the analysis given by Brook and Lantham[18] would apply to this case, except that the modulation of the reflected signal from the low energy laser would be at the frequency ω of equation (60), rather than a distribution of different frequencies. By observing the reflected signal of the low energy laser and sweeping through a range of values of ω, one could obtain some estimate of the distribution of particle size.

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[18] *M. Brook and D. J. Lantham, J. Geophys. Res., 73 (1968), 7137.*

[19] *D. E. Wortman, Possible Use of Two Laser Beams to Determine Particle Size Distribution, Harry Diamond Laboratories HDL-TR-1878 (January 1979).*

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